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Sensitivity of Modeled Number Concentrations to the Representation of New Particle Formation and Particle Emissions in Chemical Transport Models

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INTRODUCTION

Uncertainties in new particle formation (NPF) and emissions rates for particle number yield large uncertainties in modeled number concentrations (N). In this study, sensitivity studies were performed with a variety of approaches to NPF and several number emission rates using the Community Multiscale Air Quality (CMAQ) regional-scale model over all of the continental U.S with 60-km resolution for July 2004..

MODEL DESCRIPTION

NPF approaches included binary H_2SO_4–H_2O nucleation (Vehkamaki *et al.*, 2002; V02), ternary H_2SO_4–H_2O–NH_3 nucleation (Napari *et al.*, 2002; N02), and nucleation by ion-ion recombination (Turco *et al.*, 1998; NIIR). Newly-formed particles entered the Aitken mode at 3 nm diameter. The Kerminen and Kulmala (2002) parameterization for the ratio F_{KK02} of the NPF rate J_p at d_{npf} (3 nm in this work) to the nucleation rate J (1 nm) was included in some model variants. Empirical rates for the formation of 3–4 nm particles under clean conditions of the form $J_p = K[H_2SO_4]^n$, n=1, 2 (Eisele and McMurry, 1997; EM97) were also included. After calculating $J_p = F_{KK02} J$ or $J_p = J$, the (constant) J_p over the 15-min time step was limited by the available H_2SO_4.

RESULTS

Overall model sensitivities were examined using average J_p and N over the domain and simulated period ($J_{p,\,ave}$ and N_{ave}). The basecase J_p was the sum of the N02 and NIIR rates reduced by the factor F_{KK02}. Key results follow.

1. Using $J_p = F_{KK02} J$ rather than $J_p = J$ reduced $J_{p,\,ave}$ by a factor of ~3 and N_{ave} by a factor of ~2, even though the conversion is only to slightly larger (3-nm) size.
2. Replacing the ternary rate with the binary rate decreased $J_{p,\,ave}$ by a factor ~0.05 but decreased N_{ave} by only ~0.4. Binary nucleation was usually negligible in the boundary layer (PBL); above the PBL the extent of binary nucleation approached that of ternary nucleation because of lower temperatures and $[NH_3]$.
3. When the binary nucleation rate was used, inclusion of NIIR increased both $J_{p,\,ave}$ and N_{ave} only modestly.
4. The NPF rate of the average of the two n = 2 curves of Fig. 7 of EM97 yielded $J_{p,\,ave}$ intermediate between that of the binary and ternary parameterizations in the PBL, and comparable to the binary $J_{p,\,ave}$ above the PBL.
5. Reducing the mean diameter of emitted particles by a factor of two (for fixed mass emission rate) for both the Aitken and accumulation modes gave a modest decrease in both $J_{p,\,ave}$ and N_{ave}. This resulted from increased surface area and reduced NPF when emitted mass is apportioned into smaller particles. Increasing the emitted mean diameter increased $J_{p,\,ave}$ and N_{ave}.
6. Particle transfer from the Aitken to the accumulation mode is necessary in modal models to maintain distinct modes. Transfer is not governed by a physical process, and for large NPF rates this introduces a substantial uncertainty in accumulation mode number.
7. When the NPF rate is large, the subsequent reduction of J_p due to limited H_2SO_4 was also large with the 15-min time step. Nucleation and condensation are partially operator-split in CMAQ. When its binary nucleation rate is replaced by the N02 ternary rate, H_2SO_4 consumption is biased in favor of NPF.
8. In the Aitken mode, average NPF and coagulation rates nearly balanced. In the accumulation mode, number emissions and intermodal transfer were balanced by wet deposition.

CONCLUSIONS

New particle formation dominates uncertainties in Aitken mode number concentration, whereas conversion of mass emission rates to number emission rates dominates uncertainties in accumulation mode number.

Keywords: *New particle formation*

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